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(54) **Heat-curable silicone rubber compositions having resistance to engine oils and coolants**

(57) A heat-curable silicone rubber composition is disclosed having improved resistance to engine oils and coolants, including extended-life coolants and synthetic motor oils. Said composition comprises:

- (A) 100 parts by weight of an organosiloxane polymer base comprising a mixture of an organosiloxane polymer, containing at least 2 silicon-bonded alkenyl groups in each molecule and 1 to 65 weight percent of a reinforcing silica filler,
- (B) a curing component in an amount sufficient to

cure the composition when heated, and

(C) an effective amount of at least one metal salt additive selected from monobasic alkali metal phosphates, alkali metal oxalates, alkali metal tartrates, alkali metal tetraborates, alkali metal phthalates and alkali metal citrates; dibasic metal phosphates where the metal is sodium, potassium, calcium or magnesium; metal acetates, where the metal is sodium, potassium, calcium or magnesium; metal sulfates where the metal is sodium, potassium, calcium, magnesium, aluminum or zinc; and metal carbonates where the metal is sodium, potassium, calcium, magnesium, aluminum or zinc.

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Description

[0001] The present invention is a heat-curable silicone rubber composition with high resistance to degradation by engine oils and coolants and, in particular, resistance to synthetic engine oils and extended-life engine coolants.

[0002] Gaskets and packing materials formed from silicone rubber frequently suffer from poor resistance to hot hydrocarbon oils, for example, engine oil and gear oil, and from a poor resistance to radiator coolants. As a consequence, oil and coolant leaks may develop during the long-term use of silicone rubber as gasket materials in such applications.

[0003] Synthetic engine oils have as major components poly-alpha-olefins and esters which may break down to acids. Synthetic engine oils may also contain lesser amounts of additives such as oxidation inhibitors, rust inhibitors, anti-wear and extreme pressure agents, friction modifiers, detergents and dispersants, pour-point depressants, viscosity improvers and foam inhibitors. These components of synthetic oils may interact with silicone rubber differently than with hydrocarbon oils, to adversely impact the sealing properties of the rubber.

[0004] Similarly, extended-life coolants may contain organic acids, such as aliphatic monobasic acids, hydrocarbyl dibasic acids and the alkali metal, ammonium or amine salts of monobasic acids or hydrocarbyl dibasic acids, as components which interact with silicone rubber, in addition to conventional additives such as ethylene glycol, water and corrosion inhibitors, see for example, U.S. Patent No. 4,647,392. These acids and salts can attack or degrade the silicone rubber.

[0005] Fluorosilicone rubbers are generally known in the art for their resistance to fuel, oil, chemicals and solvents. However, fluorosilicone rubbers are relatively costly materials and not considered to be cost effective in many applications involving contact with engine oils and coolants. Therefore, there is a need to improve the performance of non-fluorinated silicone rubbers in contact with engine oils and coolants.

[0006] U.S. Patent 4,689,363 teaches compositions for room-temperature curable silicone rubber that are resistant to conventional engine oils. The compositions comprise 100 parts by weight of a hydroxy end-terminated polydiorganosiloxane having a linear molecular structure; from 1 to 25 parts by weight of an organosilicone having, in each molecule, at least two hydrolyzable groups bonded to the silicon atom or atoms; and from 1 to 50 parts of an alkali metal salt of a weak acid having a pK_a in the range from 2.0 to 12.0 at 25°C. The polyorganosiloxane has a viscosity in the range of 25 to 500 Pa·s or preferably from 1 to 100 Pa·s at 25°C.

[0007] U.S. Patent 5,013,781 claims compositions for room-temperature curable silicone rubber resistant to conventional coolants and hydrocarbon oils. A polyorganosiloxane composed of $R'_3SiO_{1/2}$ and SiO_2 units or $R'_3SiO_{1/2}$, R'_2SiO and SiO_2 units is included at 1 to 50 weight parts in a composition containing 100 parts polydiorganosiloxane, 5 to 300 weight parts inorganic filler, 0.1 to 10 weight parts alkoxysilane adhesion promoter and a ketoxime silicon compound crosslinker. Koshii et al. teach that the polyorganosiloxane functions in combination with the alkoxysilane adhesion promoter to improve the hydrocarbon oil and coolant (chemical) resistance of room-temperature curable silicone rubber. The molar ratio of the $R'_3SiO_{1/2}$ to SiO_2 in the polyorganosiloxane must be from 0.5:1 to 1.5:1. The polydiorganosiloxane is a flowable polymer and has a viscosity within the range of 0.0001 to 0.1 m²/s at 25°C., and the chain terminals contain a silicon-bonded hydroxyl group or a silicon-bonded hydrolyzable group.

[0008] The solutions by the above patents do not address the need for silicone compositions that are used in contact with synthetic engine oils or extended-life coolants. Furthermore, while these approaches are useful for room-temperature, vulcanizable compositions, they do not address the additional need for heat-curable silicone compositions with improved resistance to coolants and oils. Heat-curable silicone rubbers are used in applications that experience much higher stress and are possibly exposed to higher temperatures and pressures or harsher chemical environments. For example, a heat-curable silicone rubber is normally used in applications requiring tensile strengths of from 60 to 106 kg / cm², while room-temperature, vulcanizable compositions are more typically employed at lower tensile strengths of 10 to 35 kg/cm². More particularly, heat cured silicone rubbers are used in engine and coolant system applications where such systems are under heat or pressure and the properties such as compression set or compression stress relaxation are of concern. Furthermore, heat-curable silicone rubbers often have larger cross-sectional areas exposed to chemical agents, compared to room-temperature, vulcanizable compositions, which are typically used as thin gaskets. This additional cross-sectional area exposes more surface to chemical attack. Further, as gaskets are exposed to temperature cycling, they tend to swell in oil or coolant when heated and shrink when cooled. Thus, increased cross-sectional areas of heat-cured silicone rubber gaskets significantly multiplies the chemical exposure of the entire gasket.

[0009] Therefore, a non-fluorinated, heat-curable silicone rubber composition is needed that is resistant to standard and extended-life engine coolants or to standard hydrocarbon and synthetic motor oils.

[0010] This invention is a heat-curable silicone rubber composition comprising:

- (A) 100 parts by weight of an organosiloxane polymer base comprising a mixture of an organosiloxane polymer containing at least 2 silicon-bonded alkenyl groups in each molecule and 1 to 65 weight percent of reinforcing silica filler,
- (B) an amount of a curing component sufficient to cure the composition when heated, and

(C) an effective amount of at least one metal salt additive selected from

monobasic alkali metal phosphates, alkali metal oxalates, alkali metal tartrates, alkali metal tetraborates, alkali metal phthalates and alkali metal citrates;
 5 dibasic metal phosphates where the metal is selected from sodium, potassium, calcium and magnesium;
 metal acetates where the metal is selected from sodium, potassium, calcium and magnesium;
 metal sulfates where the metal is selected from sodium, potassium, calcium, magnesium, aluminum and zinc;
 and
 10 metal carbonates where the metal is selected from sodium, potassium, calcium, magnesium, aluminum and zinc.

[0011] The compositions of this invention provide superior compression set and compression stress relaxation results over prior art silicone compositions.

[0012] Component A, the organosiloxane polymer base, comprises a mixture of an organosiloxane polymer with a reinforcing silica filler. The organosiloxane polymer has the average formula of $R_aSiO_{(4-a)/2}$. In said formula, R is selected from substituted or unsubstituted monovalent hydrocarbon groups and is exemplified by alkyl groups such as methyl, ethyl and propyl; alkenyl groups such as vinyl, allyl, butenyl and hexenyl; aryl groups such as phenyl; and aralkyls such as 2-phenylethyl. The subscript "a" is a value from 1.95 to 2.05.

[0013] This organosiloxane polymer has at least 2 silicon-bonded alkenyl groups in each molecule. The alkenyl groups are bonded in pendant positions, at the terminal positions or at both positions. The molecular structure of this polymer generally has a degree of polymerization (dp) in the range of from 200 to 20,000, and preferably a dp of 1000 to 20,000. This dp range includes polymers which are thick, flowable liquids as well as those that have a stiff, gum-like consistency. Our organosiloxane polymer can be a homopolymer, a copolymer or a mixture of such polymers. The siloxy units comprising said polymer are exemplified by dimethylsiloxo, vinylmethylsiloxo and methylphenylsiloxo. The molecular terminal groups in the polymer are trimethylsiloxo or vinyltrimethylsiloxo groups.

Representative organosiloxane polymers are exemplified by vinyltrimethylsiloxo-endblocked dimethylsiloxane-vinylmethylsiloxane copolymer, vinyltrimethylsiloxo-endblocked polydimethylsiloxane, vinylmethylhydroxysiloxo-endblocked dimethylsiloxane-vinylmethylsiloxane copolymer and vinyltrimethylsiloxo-endblocked dimethylsiloxane-methylphenylsiloxane-vinylmethylsiloxane copolymer. A preferred polymer is a vinyltrimethylsiloxo-terminated polydimethylsiloxane gum comprising 0.142 mole percent of vinylmethylsiloxane units and exhibiting a plasticity of 1.38 - 1.62 mm (55 - 65 mils) based on ASTM D926.

[0014] Our base also contains a reinforcing silica filler to provide increased mechanical properties for the product present heat cured silicone rubber composition. The filler is any silica filler that is known to reinforce organosiloxanes and is preferably selected from particulate, fumed or precipitated forms of silica and silica aerogels which have a specific surface area of at least 50 m²/g, and preferably 150 to 400 m²/g. The filler is typically added at a level of 1 to 65 weight percent of the organosiloxane polymer base, and preferably is 10 to 65 weight percent of the base.

[0015] It is preferred to treat the reinforcing silica filler to render its surface hydrophobic, as typically practiced in the art. This is accomplished by reacting the reinforcing silica filler with a liquid organosilicon compound that contains silanol groups or hydrolyzable precursors of silanol groups. Useful as filler treating agents, also referred to as anti-creeping agents or plasticizers in the art, include low molecular weight hydroxy- or alkoxy-terminated polydiorganosiloxanes, including α,ω -silanediols, hexaorganodisiloxanes, cyclodimethylsiloxanes and hexaorganodisilazanes.

[0016] In addition to organosiloxane polymer and reinforcing silica filler, our organosiloxane polymer base may also contain other additives, for example heat stability additives, anti-structure agents, pigments and extending or semi-reinforcing fillers. Examples of additives include diatomaceous earths, ground quartz, zinc oxide, calcium carbonate, titania and magnesium oxide. The proportion of such additional fillers will depend on the physical properties and other characteristics desired in the final elastomer. Generally, the additional fillers are present in a proportion of from 10 to 150 parts by weight of the organosiloxane polymer.

[0017] The curing component (B) is selected from any of the well-known curing systems for the silicone rubber art. For example, the curable silicone rubber compositions of this invention are cured to the elastomeric state by exposure to electron beams, ultraviolet rays, electromagnetic waves or heat. When heat is used, an organic peroxide curing agent may be employed. Examples of suitable curing agents include 2,5-dimethyl-2,5-di(tert-butylperoxy) hexane, 2,2-bis(t-butylperoxy)-p-diisopropyl-benzene, 1,1-bis(t-butylperoxy)-3,3,5-trimethylcyclohexane, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexyne-3, di-t-butylperoxide, benzoyl peroxide, p-chlorobenzoyl peroxide, dicumyl peroxide, tertiary butyl peracetate, tertiary butyl perbenzoate, monochlorobenzoyl peroxide, 2,4-dichlorobenzoyl peroxide and tertiary butyl cumyl peroxide. A preferred organic peroxide curing agent is 2,5-dimethyl-2,5-di(tert-butylperoxy) hexane.

[0018] Another heat curable system which is applicable herein is where the curable silicone rubber composition is cured by crosslinking the organosiloxane with an organohydrogensiloxane crosslinker in the presence of a platinum-group, metal-containing catalyst. Such a crosslinker usually contains an average of at least two silicon-bonded hydro-

gen atoms per molecule and no more than one silicon-bonded hydrogen atom per silicon atom. The remaining valences of the silicon atoms are satisfied by divalent oxygen atoms or by monovalent hydrocarbon radicals comprising one to seven carbon atoms. Acceptable monovalent hydrocarbon radicals are, for example, alkyls such as methyl, ethyl, propyl, tertiary butyl and hexyl; cycloalkyls such as cyclohexyl; and aryls such as phenyl or tolyl. The platinum-group, metal-containing catalyst is any such catalyst which is known to catalyze the reaction of silicon-bonded hydrogen atoms with silicon-bonded vinyl groups. By platinum-group metal, we mean ruthenium, rhodium, palladium, osmium, iridium and platinum.

[0019] Component (C) is an effective amount of a metal salt additive selected from at least one of the group consisting of monobasic alkali metal phosphates; alkali metal oxalates; alkali metal tartrates; alkali metal tetraborates; alkali metal phthalates; alkali metal citrates; and dibasic metal phosphates where the metal is selected from the group consisting of sodium, potassium, calcium and magnesium. Also included are metal acetates, where the metal is selected from the group consisting of sodium, potassium, calcium and magnesium. Further covered are metal sulfates where the metal is selected from the group consisting of sodium, potassium, calcium, magnesium, aluminum and zinc. And finally provided for are metal carbonates where the metal is selected from the group consisting of sodium, potassium, calcium, magnesium, aluminum and zinc.

[0020] Without being bound to any particular theory, we believe that our metal salt additives which impart resistance to coolants and synthetic oils in the present invention act in a manner analogous to salts which are present in an aqueous buffer system. Aqueous buffer systems are well known in chemical arts and are characterized by their ability to resist pH changes when diluted or when various amounts of acid or base are added. An article describing how such buffer systems work is Peters, Hayes, and Hieftje, "Aqueous Acid-Base Reactions," Chemical Separation and Measurements, W. B. Saunders Company (1974), pp. 100-112. Silicone rubber is more stable in a chemical environment where the pH of a system is at neutral or near-neutral. In the present invention, metal salts which are used in conventional buffer solutions to maintain a pH in a range of 3 to 8 are useful.

[0021] For water-soluble metal salt additives, a pKa between 3 and 8 is useful. For additives with limited solubility, the salt should be capable of increasing the pH of a dilute aqueous acidic solution from a pH below 3 to a pH between 3 and 8, or reducing a solution with a pH of greater than 8 to a pH between 3 and 8.

[0022] By effective amount of a metal salt additive is that amount which provides a heat-curable silicone rubber composition, retaining greater than 35 percent of the sealing force when exposed to long life coolant for 6 weeks in a compression stress relaxation (CSR) test, or retaining more than 10 percent of sealing force while still exhibiting a compression set of at most 40 percent after 6 weeks exposure in synthetic motor oil. Compression stress relaxation methods are more fully described below in the examples. A typical additive amount will be in the range of 0.5 to 20 weight parts of salt for 100 weight parts of organosiloxane polymer base.

[0023] When resistance to extended-life coolants is desired, our metal salt additive is preferably an alkali metal monobasic salt selected from the group consisting of phosphates, oxalates, tartrates, tetraborates, phthalates, citrates, acetates, sulfates and carbonates. This salt is usually sodium or potassium, though not limited to these metals. A salt for this purpose is monosodium phosphate. When monosodium phosphate, NaH_2PO_4 , is used, the amount is typically 0.5 to 7.5 parts by weight of monosodium phosphate per 100 parts by weight of the organosiloxane base. Amounts below 0.5 parts by weight of this salt per 100 weight parts base may give less than desired resistance to said coolant. Surprisingly, the effectiveness is also reduced for amounts over 7.5 parts by weight of salt per 100 weight parts of base.

[0024] When resistance to synthetic motor oil is desired, our metal salt additive is a dibasic metal salt selected from the group consisting of dibasic metal phosphates, where said metal is selected from the group consisting of sodium, potassium, calcium and magnesium; dibasic alkali metal oxalates; dibasic alkali metal tartrates; dibasic alkali metal tetraborates; dibasic alkali metal phthalates; dibasic alkali metal citrates; metal acetates, where the metal is selected from the group consisting of sodium, potassium, calcium, magnesium; metal sulfates where the metal is selected from the group consisting of sodium, potassium, calcium, magnesium, aluminum and zinc; and metal carbonates where the metal is selected from the group consisting of sodium, potassium, calcium, magnesium, aluminum and zinc. A preferred salt is disodium phosphate, Na_2HPO_4 . A preferred amount for disodium phosphate is from 0.5 to 20 weight parts of said salt per hundred weight parts of organosiloxane base. Concentrations of disodium phosphate below 0.5 weight part per 100 parts of organosiloxane base do not provide the desired resistance to motor oil. At concentrations above 20 parts, the physical properties of the heat-curable silicone rubber product when cured are not desirable.

[0025] A combination of metal salt additives can also be used to provide resistance to both said coolants and oils. When a combination of metal salt additives is used, the total amount of salts will not exceed 20 parts of the combined weights of the metal salt additives per hundred weight parts of organosiloxane polymer base. As above, the physical properties of the heat-curable silicone rubber product are not desirable above this upper limit of 20 weight parts.

[0026] The compositions of the present invention are prepared by any convenient procedure. The organosiloxane polymer base and the additive (C) are readily mixed together with any other ingredients or extending fillers, with sufficient heat and shear to achieve a uniform base. In a preferred method, a high consistency organosiloxane polymer base is first prepared by mixing organosiloxane polymer with reinforcing filler and then with any extending fillers or

other ingredients. Next, the metal salt additive is incorporated into said base, using any mixing device, such as a two-roll mill. The extending filler amount to be added is normally 1 - 100 parts by weight of extending filler per 100 parts by weight of the organopolysiloxane polymer. Suitable extending fillers are represented by crushed or pulverized quartz, diatomaceous earth, calcium carbonate, zinc carbonate, magnesium oxide, iron oxide, zinc oxide, titanium oxide, powdered mica, carbon black, graphite and glass fibers or microspheres.

Examples

Materials

[0027] The following materials were used for testing sample compositions:

OIL 1 is MOBIL™ 1 Advanced Formula, 5W-30 synthetic motor oil, API Service SJ/CF, from Mobil Oil Corporation, Fairfax, VA. This oil is formulated from poly-alphaolefin and ester, together with an additive package.

OIL 2 is MOBIL™ 5W-30 High Performance, API Service SJ, from Mobil Oil Corporation, Fairfax, VA.

COOLANT 1 is Havoline DEX-COOL® Extended Life Anti-Freeze/Coolant, Code 7995, manufactured by Texaco Lubricants Company, Houston, TX. This is a single-phase, ethylene glycol engine coolant, which incorporates organic acid corrosion inhibitor carboxylate technology, mixed 50/50 by volume, with deionized water.

Test methods

[0028] All silicone rubber samples were compounded on a 2-roll mill. Standard test slabs were molded 10 minutes at 171°C. following ASTM procedures. The following standard tests were then conducted and the test materials were evaluated: Durometer, Shore A-2 (ASTM D 2240); tensile strength, elongation and modulus (ASTM D 412); and compression set (ASTM D395B) for 22 hours at 177°C.

[0029] A hot oil compression set test was used herein as an indicator of gasket performance. Square cross-section o-rings (25 mm O.D. x 17 mm I.D. x 4-mm thickness) were compression-molded for 15 minutes at 171°C. These o-rings were then placed between two parallel steel plates with a central hole and compressed 20% using shims or corner bolts, while submerged in motor oils at 150°C. Every two weeks, the fixtures containing these o-rings were cooled to room temperature; removed from the oil; disassembled and measured for height loss after stabilizing for a day. Compression sets were thereafter calculated from this data. The oil was changed at each inspection interval before restarting the test using the same test specimens. Testing was continued for several weeks, or until 100% compression set occurred. All results are reported as the percentage of compression set observed in Table 2.

[0030] A compression stress relaxation (CSR) test was also used to test gasket performance. This method is more fully described in ASTM D6147, the test procedure for "Rubber, Vulcanized Determination of Force Decay (Stress Relaxation) in Compression". The test was conducted using 19 mm O.D. x 12.5 mm I.D. o-rings, die-cut from slabs molded 10 minutes at 171°C. Oil testing was done with 2 mm thick o-rings and coolant testing was done with 3 mm thick o-rings. The rings were placed in Jamak™ CSR Test Fixtures, compressed 25% and then measured for initial sealing force one-half hour after set-up. The test fixtures were next placed in containers and covered with test fluids. Thereafter, the containers were placed in ovens at 150°C. for oil testing. For coolant testing, the fixtures were placed in coolant in pressure vessels to prevent water evaporation at 125°C. test temperatures. Sealing force measurements were taken after one day and three days, and then weekly throughout the rest of the tests. The fixtures were removed from the hot test fluids and cooled to room temperature (2 hours) before force measurements were taken using a Shawbury-Wallace™ Testing Tower to measure sealing forces over time. Oils were changed weekly, whereas the same coolant mixture was used throughout the test. Percentage sealing force retention was subsequently determined from the force measurements.

[0031] For CSR oil testing, Mobil™ 1 AF synthetic oil was used. Both Mobil™ 1 and Mobil™ HP 5W-30 oils were used for hot oil compression set testing.

[0032] The base material (Base 1) used in the examples was prepared in a dough mixer. It contained the following materials: 65-70 weight percent of a polyvinylmethyl-dimethylsiloxane gum co-polymer containing 0.1 mole percent of a vinylidimethyl-endblocked-polyvinylmethylsiloxane; 4-6 weight percent of a hydroxyl-endblocked polyphenylmethylsiloxane process aid; 0.5 to 1.5 weight percent of a polyvinylmethylsiloxane process aid; 20-35 weight percent of a fumed silica having a surface area of 200 m²/g; and a trace amount of ammonia. These ingredients were mixed and heated for several hours at elevated temperature to make a shelf-stable silicone rubber base.

[0033] The materials tested were next compounded using a standard laboratory 2-roll mill. The following ingredients were used: (Base 1) base material; (Filler) 5 micrometer Min-U-Sil®, U.S. Silica, Berkley Springs, WV; (Pigment) black pigment (50% carbon black/50% polyvinylmethylsiloxane gum); (Curing agent 1) 2,5-Dimethyl-2,5-di(tert-butylperoxy) hexane (mixed at 45% in inert carriers); (Additive 1) monosodium phosphate, and (Additive 2) disodium phosphate.

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Quantities used are given in Table 1 and test results are reported in Table 2. The results show that in the coolant 74% of the sealing force was retained in the CSR test when the curable sample contained 2.56 weight parts of monosodium phosphate. This demonstrates a significant improvement in sealing force retention over samples with no monosodium phosphate. Surprisingly, when 8.11 parts of monosodium phosphate were used, there was not a significant difference in the CSR test results compared to samples with no monosodium phosphate.

[0034] The results also show that samples with disodium phosphate showed lower hot oil compression set values and higher percent sealing force retention in CSR tests as compared to samples without disodium phosphate.

Table 1.

Test formulations						
<u>Compounds</u>	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>	<u>E</u>	<u>F</u>
Base 1, wt. Parts	100	100	100	100	100	100
Filler, wt. Parts	20.0	21.6	28.9	21.6	20.5	21.3
Pigment, wt. Parts	1.00	1.08	1.04	1.08	1.02	1.06
Curing agent, wt. Parts	1.00	1.08	1.04	1.08	1.02	1.06
Additive 1, wt. Parts	0.00	0.00	0.00	8.11	2.56	1.33
Additive 2, wt. Parts	0.00	8.11	3.90	0.00	0.00	5.33

Table 2.

Test results						
<u>Compounds</u>	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>	<u>E</u>	<u>F</u>
Durometer, Shore A-2, mils	1.60	1.65	1.62	1.62	1.62	1.62
Tensile Strength, MPa	8.4	4.8	5.6	7.4	7.7	4.8
Elongation, %	285	165	185	275	275	160
Modulus at 100% Elongation, MPa	2.8	2.8	2.9	2.8	2.8	2.8
Hot Air Compression Set after 22 h at 177°C., %	6	11	11	23	16	13
Hot Oil Compression Set after 4 weeks in Oil 1 at 150°C., %	100	29	25	94	100	33
Hot Oil Compression Set after 4 weeks in Oil 2 at 150°C., %	52	22	17	52	93	17
Compression Stress Relaxation after 6 weeks in Oil 1 at 150°C., sealing force retained, %	0	18	19	0	16	17
Compression Stress Relaxation after 6 weeks in Coolant 1 at 125°C., sealing force retained, %	22	5	21	12	74	15

Claims

1. A heat-curable silicone rubber composition comprising:

(A) 100 parts by weight of an organosiloxane polymer base comprising an organosiloxane polymer containing at least 2 silicon-bonded alkenyl groups in each molecule and 1 to 65 weight percent of a reinforcing silica filler, (B) an amount of a curing component sufficient to cure the composition when heated, and (C) an effective amount of at least one metal salt additive selected alkali metal citrates, alkali metal oxalates, monobasic alkali metal phosphates, alkali metal phthalates, alkali metal tartrates and alkali metal tetraborates;

dibasic metal phosphates where the metal is selected from sodium, potassium, calcium and magnesium; metal acetates, where the metal is selected from sodium, potassium, calcium and magnesium; metal sulfates where the metal is selected from sodium, potassium, calcium, magnesium, aluminum and zinc; and metal carbonates where the metal is selected from sodium, potassium, calcium, magnesium, aluminum and zinc.

2. The composition according to claim 1,

wherein said organosiloxane polymer has the average formula of $R_a\text{SiO}_{(4-a)/2}$ and R is a substituted or unsubstituted monovalent hydrocarbon group and "a" is a value from 1.95 to 2.05.

3. The composition according to claim 1 or 2,
wherein said organosiloxane polymer base further comprises 1 to 100 parts by weight of an extending filler, per 100 parts by weight of the polyorganosiloxane polymer.
4. The composition according to any of claims 1 to 3, wherein Component (B) comprises an organohydrogensiloxane crosslinker.
5. The composition according to any of claims 1 to 4, wherein component (B) comprises a platinum-group, metal-containing catalyst.
6. The composition according to any of claims 1 to 5, wherein component (C) comprises 0.5 to 20 weight parts of metal salt additive per 100 weight parts of organosiloxane polymer base.



European Patent
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EUROPEAN SEARCH REPORT

Application Number
EP 99 30 2914

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	EP 0 752 450 A (DOW TORAY) 8 January 1997 (1997-01-08)	1-5	C08K3/26 C08K5/098 C08L83/04
Y	* page 5, line 41,42 * * page 6, line 12 - line 14 * * examples * * claim 1 *	1,6	
D,Y	--- US 4 689 363 A (Y. INOUE ET AL.) 25 August 1987 (1987-08-25) * column 1, line 8 - line 12 * * column 3, line 60 - column 4, line 27 * * column 5, line 19 - line 28 * * claims * -----	1,6	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C08K C08L
The present search report has been drawn up for all claims			
Place of search BERLIN		Date of completion of the search 20 July 1999	Examiner Hoepfner, W
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>			

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**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

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This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report.
The members are as contained in the European Patent Office EDP file on
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For more details about this annex : see Official Journal of the European Patent Office, No. 12/82